



Greenhouse gas benefits from direct chemical recycling of mixed plastic waste

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ABSTRACT

Dealing with heterogeneous plastic waste – i.e., high polymer heterogeneity, additives, and contaminants – and lowering greenhouse gas (GHG) emissions from plastic production requires integrated solutions. Here, we quantified current and future GHG footprints of direct chemical conversion of heterogeneous post-consumer plastic waste feedstock to olefins, a base material for plastics. The net GHG footprint of this recycling system is -0.04 kg CO₂-eq./kg waste feedstock treated, including credits from avoided production of virgin olefins, electricity, heat, and credits for the partial biogenic content of the waste feedstock. Comparing chemical recycling of this feedstock to incineration with energy recovery presents GHG benefits of 0.82 kg CO₂-eq./kg waste feedstock treated. These benefits were found to increase to 1.37 kg CO₂-eq./kg waste feedstock treated for year 2030 when including (i) decarbonization of steam and electricity production and (ii) process optimizations to increase olefin yield through carbon capture and utilization and conversion of side-products.

1. Introduction

Various emerging waste treatment technologies aim to increase the environmental performance of plastics and plastic waste management. Existing plastic waste management schemes rely mostly on landfilling and incineration. It was estimated that up to 2015, approximately 6.3 Gt of plastic waste was generated globally since the advent of plastics, of which 79% accumulated in landfills or the natural environment, 21% was incinerated, and only 9% was collected for recycling (Geyer et al., 2017). Mechanical recycling is currently the only plastic waste recycling option deployed at scale, which entails technologies to process and recirculate plastic waste without significantly altering the chemical

structure of the material. Polymer heterogeneity, (non-polymer) additives and contaminants in plastic waste are all limiting factors in deployment of mechanical recycling. Furthermore, low product quality and higher costs of recycled plastics compared to virgin plastics impede market competitiveness (Brouwer et al., 2020). However, favouring virgin plastic production over recycling clearly comes with environmental consequences: Greenhouse gas (GHG) emissions from production, incineration, and degradation in landfills contribute to global warming. New recycling technologies are therefore developed to provide integrated solutions for plastic waste that is currently not recirculated.

Chemical recycling – also referred to as tertiary recycling or

Abbreviations: BB, biogas boiler; BECCS, bioenergy with carbon capture and storage; BL, baseline; CCU, carbon capture and utilization; CHP, combined heat and power; EB, electric boiler; ESP, emission saving potential; GHG, greenhouse gas; GWP, global warming potential; HVC, high value chemical; LCA, life cycle assessment; LCI, life cycle inventory; LHV, lower heating value; MPL, market penetration level; MSW, municipal solid waste; MTO, methanol-to-olefins; RCP, representative concentration pathway; SSP, shared socioeconomic pathway; T.EN, Technip Energies; TRL, technology readiness level.

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feedstock recycling – is a category of novel technologies in which plastic waste is transformed through the breaking of chemical bonds. This can be through decomposition of polymers to monomers or conversion of polymers to a liquid or gaseous mixture of chemicals used as feedstock in polymer production (Vollmer et al., 2020). Of the currently available technologies ($n = 50$) listed in a recent survey by Closed Loop Partners (2021), only eight reached the maturity stage of commercial growth. Most chemical recycling technologies are at an early-commercial or pre-commercial stage and require further development in the upcoming years (see section 1 of Supplementary Materials 1).

Chemical recycling technologies of the conversion type were found to be the most mature (Closed Loop Partners, 2021), with common technologies being pyrolysis and gasification. Pyrolysis is performed in absence of oxygen at temperatures of around 300–600 °C (Broeren et al., 2019), producing pyrolysis oil. After processing, this pyrolysis oil can be used to substitute naphtha in conventional petrochemical processes to produce virgin plastics. Gasification is performed with a gasifying agent (e.g., oxygen, steam, air) at higher temperatures of 700–1600 °C (Broeren et al., 2019), producing a gaseous mixture. The term gasification typically refers to the production of syngas, consisting of hydrogen gas and carbon monoxide, which is obtained at temperatures of >1100°C. To produce virgin plastics, conversions are required to olefins *via* methanol or to a naphtha substitute *via* the Fischer-Tropsch process. At temperatures around 700–800 °C, however, olefins can be obtained directly from conversion of plastic waste. Already three decades ago this was considered a promising way for feedstock recycling of polyolefinic waste (Simon et al., 1996) and it has since seen development from the lab scale to the pilot scale. The direct recycling route requires few processing steps to convert plastic waste to new plastics of virgin quality and is therefore expected to present large GHG benefits when compared to other options.

Life cycle assessment (LCA) studies of plastic waste treatment technologies generally find that chemical recycling has a GHG footprint comparable to or larger than mechanical recycling, but lower than incineration with energy recovery (BASF, 2020; Davidson et al., 2021; Jeswani et al., 2021; Meys et al., 2020; Quantis, 2020; Uijttewaall & Broeren, 2021). Feedstock characteristics can, however, limit applicability of some of these waste treatment options. In mechanical recycling and pyrolysis, impurities and contaminants easily get transferred into products, which hampers further processing. Furthermore, these LCA studies demonstrated that it is relevant to specifically account for which conventional product will be replaced. For example, net GHG footprints of chemical recycling could be higher than that of incineration for scenarios where credits from plastic incineration are large due to replacement of fossil fuels with high emission factors (Jeswani et al., 2021; Meys et al., 2020). On the other hand, chemical recycling can have lower GHG footprints than mechanical recycling when taking reduction in polymer quality into account, e.g., in open-loop recycling of polyolefins and polystyrene (Schwarz et al., 2021). Finally, GHG footprints were found to be sensitive to future trends, in particular changes in process carbon efficiency and emission intensity of electricity generation (BASF, 2020; Jeswani et al., 2021; Uijttewaall & Broeren, 2021). What is missing in these LCA studies, is comprehensive comparative assessment of GHG footprints, accounting for the combined effect of various future developments.

The aim of our study is to evaluate whether current and future chemical recycling of post-consumer mixed plastic waste provides GHG benefits when compared to incineration with energy recovery. Primary data from Synova and Technip Energies (T.EN) were used to assess direct chemical conversion of post-consumer mixed plastic waste to olefins. Changes in GHG footprints for this system were assessed by including projections of decarbonization of steam and electricity production and valorisation of side-product fuel gas and captured carbon dioxide for year 2030.

2. Materials and methods

We followed the general workflow for LCA outlined in the ISO 14040 standard (ISO, 2006), complemented with the systematic approach to assess the environmental impact of emerging technologies described in van der Hulst et al. (2020).

2.1. Goal and scope

Goal of conducting a prospective GHG footprint assessment for the Synova/T.EN system was to inform further development of this technology, by identifying GHG hotspots in the chemical recycling process and assessing the effect of expected developments up to 2030. Furthermore, the goal was to compare this technology at an emerged state to its incumbent to assess whether plastic waste treatment with the Synova/T.EN system can provide GHG benefits. The assessment followed the approach of an attributional LCA, since the aim was to evaluate the GHG footprint that can directly be associated with the chemical recycling processes under the *ceteris paribus* assumption.

Synova used a plastic-rich refuse derived fuel as feedstock for trials with their system, which was sourced from municipal solid waste (MSW) by a materials recovery facility. This feedstock contains non-plastic contaminants such as food residue, paper labels, glass, and dirt, which is typical for post-consumer mixed plastic waste. Thus, the feedstock is not purely plastic material, but also contains inert materials and biomass, which is converted to high value chemicals (HVCs) together with the plastics. Following Hanssen and Huijbregts (2019), the functional unit for the assessment was “*treatment of 1 kg feedstock*”, where the feedstock was defined as “*plastic-rich refuse derived fuel containing 60.0 wt% mixed plastics, 28.1 wt% biomass, 1.0 wt% moisture and 10.9 wt% inert materials and having a lower heating value (LHV) of 29.8 MJ/kg*”. How this composition was determined, is described in section 2. of Supplementary Materials 1. The chemical recycling process is multi-functional in that it both fulfils the function of waste treatment and the function of producing multiple recycled materials. Allocation was circumvented by applying the method of substitution, an approach commonly applied in LCAs of plastic waste recycling systems (e.g., Jeswani et al., 2021; Meys et al., 2020; Uijttewaall & Broeren, 2021), in which the system is credited for avoiding production of equivalent HVCs and side-products through conventional processes such as naphtha cracking. Assumed avoided products and their quantities are provided in Fig. 1 and Fig. 2.

The scope of the assessment was a comparative assessment of GHG footprints of chemical recycling and incineration in the Netherlands for years 2020 and 2030. Waste feedstock was modelled as originating from Dutch MSW. The composition of Dutch MSW was obtained from Rijkswaterstaat (2021) and the plastic fraction in this MSW was assumed to have a global average polymer composition as modelled by McKinsey & Company (2019) due to lack of Dutch data on polymer composition. Processes of the incineration and chemical recycling systems were modelled up to the point of obtaining usable energy and/or separated HVCs that can provide a one-on-one replacement for existing energy and HVC production in the economy, in line with Hanssen and Huijbregts (2019). The use phase of these products was excluded from the comparison based on equivalence. For the end-of-life stage, GHG savings from avoided fossil CO₂ emissions at HVC end-of-life were included to account for the difference in fossil carbon content between conventional HVCs and those derived from the plastic waste feedstock. Thus, the scope of the comparative assessment was cradle-to-grave, excluding equivalent processes in the use and end-of-life stages.

Processes were modelled to take place at the Dutch petrochemical site Chemelot, because of data availability and because the site contains infrastructure suitable for post-processing. The life cycle inventory (LCI) database ecoinvent, version 3.7, system model “allocation, cut-off by classification” (ecoinvent, 2020), was used to model background processes. Dutch datasets were selected where possible and appropriate.

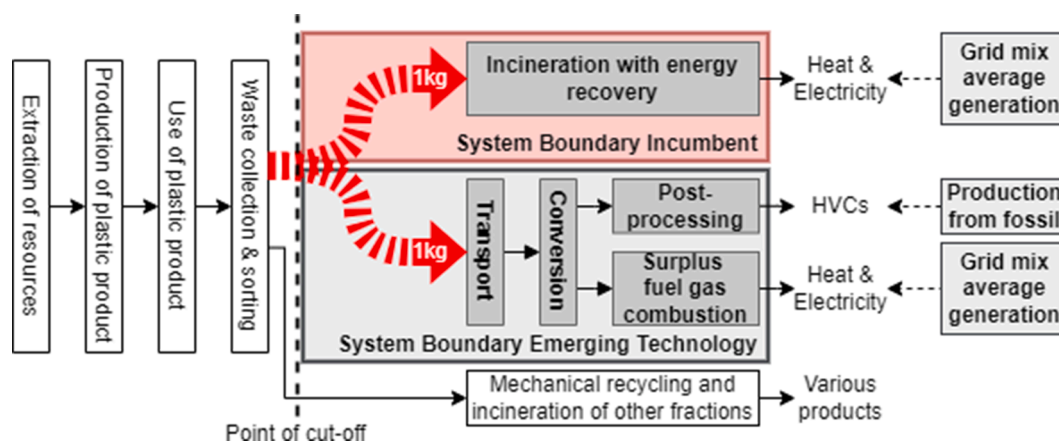


Fig. 1. Flow diagram indicating the system boundary of the modelled incumbent (incineration) and emerging (chemical recycling) product systems. Processes inside the system boundary are within the scope of the study. Processes on the right indicated avoided conventional production processes. Interventions from the life cycle of the plastic product are considered out-of-scope and are excluded up to the indicated dashed line, which is where the system boundaries of the studied product systems start. The reference flow of “1 kg feedstock treated” is indicated with the bold red dashed arrow and is identical for both product systems. HVCs: high value chemicals.

The cut-off point was placed at the end of the activity producing the recyclable material, in line with the system model description provided by ecoinvent (ecoinvent, 2022). Consequently, plastic waste at the processing facility was considered burden-free. The emission saving potential (*ESP*) from diverting plastic waste from incineration to chemical recycling was calculated using Eq. 1 from Layritz et al. (2021), adapted to our specific comparison, with a positive *ESP* indicating GHG benefits. Emissions *E* in kg CO₂-eq./kg feedstock treated for the incineration and chemical recycling processes were both calculated using Eq. 2, in which credits from obtained products were accounted for as avoided emissions from conventional production in line with Hanssen and Huijbregts (2019) (see Fig. 1 and Fig. 2).

$$ESP = \sum E_{incineration} - \sum E_{chemical\ recycling} \quad (1)$$

$$E = E_{direct} + E_{indirect} - E_{avoided\ conventional\ production} \quad (2)$$

2.2. Inventory

General approaches for compiling the inventories are described in subsequent sections and more detailed descriptions are available in section 3. of Supplementary Materials 1. Fig. 2b presents a more detailed flow chart of all relevant unit processes involved in the current (2020) product system of direct chemical recycling, with main processes highlighted in dark grey boxes as in Fig. 1. Fig. 2c presents the future (2030) direct chemical recycling system, with developments in different scenarios indicated in different colours (see 2.3 Prospective assessment). Background processes are represented using datasets from ecoinvent, except for “Fuel gas production (external)”. For this process, a dataset was created from data on fuel gas, produced and used at Chemelot (Chemelot Site Permit BV, 2020). This site produces more fuel gas than it consumes, which is currently used in combined heat and power production, but which could alternatively be used in energy processes of the studied direct chemical recycling system.

2.2.1. Conversion: Synova/T.EN system

Conversion of the feedstock to high value chemicals was modelled for the Synova/T.EN system as reported in the screening LCA by Uijtewaal and Broeren (2021). Inventory data were supplemented with primary data from Synova, obtained from trial runs in pilot scale equipment at 750 °C with an hourly throughput of 150 kg feedstock. A weighted average distance of 224 km was calculated for transportation of the waste feedstock. The Synova/T.EN system required inputs of electricity, heat from steam and various materials for processing the

product gas. These include acid, caustic, sand, and gas cleaning, gas scrubbing, and anti-foaming agents. Electricity was assumed to be supplied by the average Dutch grid mix for 2020 and steam was assumed to be generated from combustion of part of the side-product fuel gas. Steam used for heat is kept in a closed system, with condensed water being used as boiler feed water. Replenishment of water losses from this closed system were not considered. Steam used as fluidizing agent is condensed in product gas purification and is therefore not reused for steam production, but instead treated in an on-site wastewater treatment plant. Besides wastewater, ashes were the main waste to be treated, which were assumed to be landfilled. Consumption of natural gas at reactor start-up was excluded from the assessment, since it was calculated to have a negligible contribution to the overall energy consumption of the system evaluated.

2.2.2. Post-processing: separation of HVCs

The Synova/T.EN system requires integration into a host plant with infrastructure to separate the gaseous mixture of HVCs and process the individual olefins obtained. This gaseous mixture represents a “drop-in” stream that resembles HVCs already processed in the existing post-processing units. Energy consumption in post-processing was therefore derived from available data for HVC production at the Chemelot site (Oliveira & Van Dril, 2021). Credits for avoiding GHG emissions from conventional production of HVCs were calculated using datasets for average European production from PlasticsEurope present in the ecoinvent database (Althaus et al., 2007; Hirschier, 2007).

While HVCs derived from plastic waste are functionally equivalent to conventional HVCs, the former partially contains biogenic carbon from food residue and paper labels, while the latter consists solely of fossil carbon. This distinction becomes apparent at use and end-of-life for the HVCs, and products derived thereof. Assuming HVCs are converted to plastic products results in a storage period of 8.7 years, based on the weighted average lifetime of plastic products as derived from Geyer et al. (2017) (see section 3. of Supplementary Materials 1). A storage credit of -0.06 kg CO₂-eq./kg CO₂(stored) was included, based on this lifetime and assuming a conservative rotation period of 1 year for all biomass in the feedstock (Guest et al., 2013). Furthermore, GHG savings from avoided fossil CO₂ emissions at HVC end-of-life were included.

2.2.3. Surplus fuel gas combustion

After part of the side-product fuel gas is combusted in steam boilers and furnaces used in conversion and post-processing, a surplus remains which was assumed to be combusted in combined heat and power (CHP) production for external use. A thermal efficiency of 45% and electrical

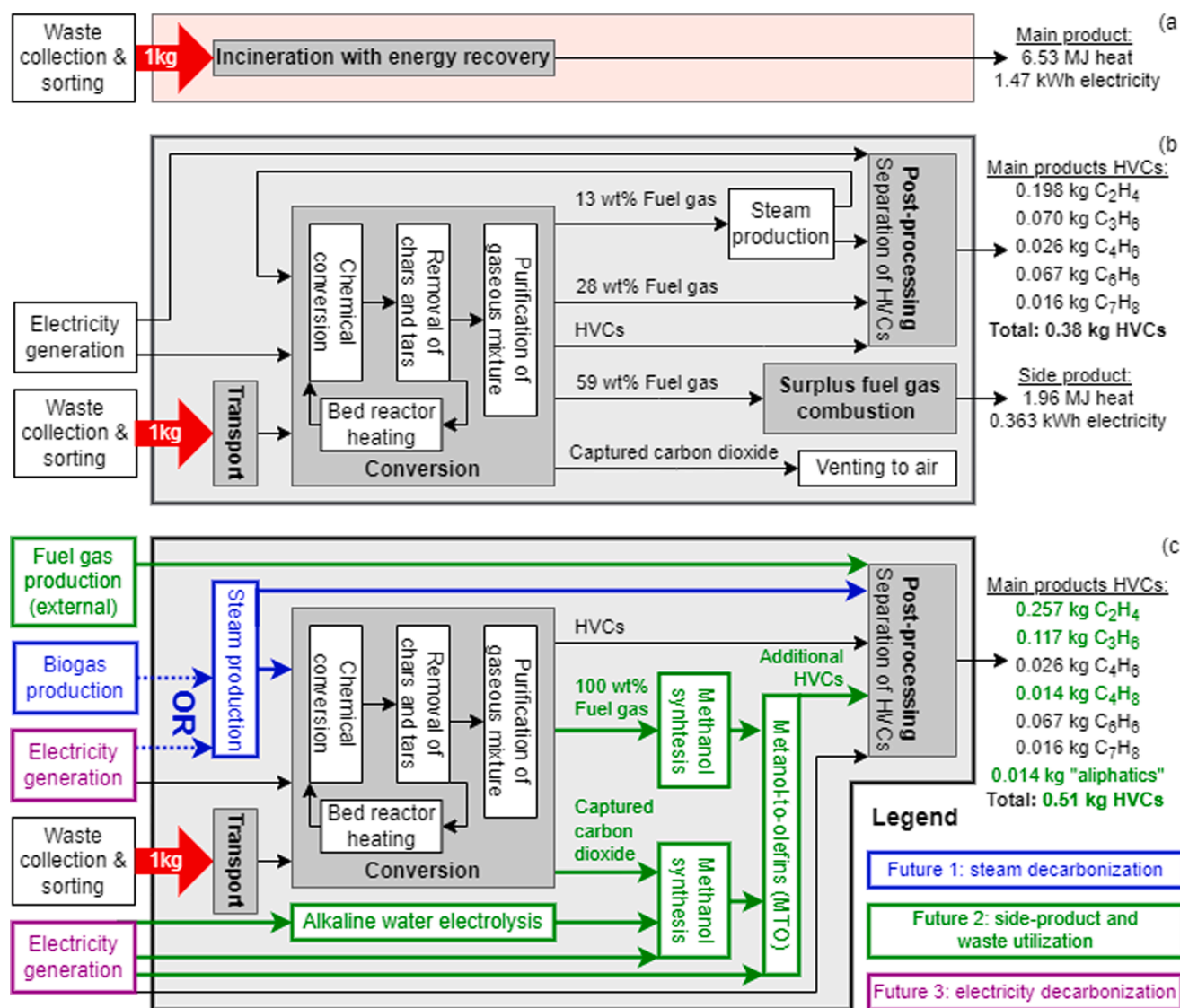


Fig. 2. Detailed flow diagram of the incumbent product system (a) and the current (b) and future (c) product system for chemical recycling. Elementary flows and small system exchanges were omitted for clarity. The reference flow of “1 kg feedstock treated” is indicated with the bold red arrow. For the future system, added or changed processes (boxes) and exchanges (arrows) are marked with different colors, as indicated in the legend, to mark the corresponding future scenario. For steam decarbonization, two alternative input exchanges were considered, which are marked with dashed arrows: either electricity or biogas is considered as energy source, but not both. C₂H₄: ethylene; C₃H₆: propylene; C₄H₆: 1,2-butadiene; C₄H₈: butene; C₆H₆: benzene; C₇H₈: toluene; HVCs: high value chemicals.

efficiency of 30% were based on the Swentibold CHP plant at the Chemelot site [Oliveira and Van Dril \(2021\)](#). Credits to the direct chemical recycling process were included for avoided Dutch market average production of high voltage electricity and district or industrial heat.

2.2.4. Incineration: incumbent waste treatment system

Theecoinvent dataset for “incineration of waste plastic, mixture” in market region “rest of world” ([Doka, 2013](#)) was used to assess GHG footprints from incineration of the feedstock. The biosphere flow “Carbon dioxide, fossil” to air was multiplied by 0.7664 to account for 23.36% of carbon in the feedstock being of biogenic origin, since carbon dioxide emissions of biogenic origin have a characterization factor of 0 kg CO₂-eq./kg. Information on waste incineration facilities in the Netherlands was used to calculate average electrical and thermal conversion efficiencies ([Rijkswaterstaat, 2020](#)). Again, credits were included for avoided Dutch market average production of electricity and heat.

2.3. Prospective assessment

The prospective LCA framework in [van der Hulst et al. \(2020\)](#) was applied to the inventory for the 2020 chemical recycling system to assess changes in the GHG footprint for year 2030 as a result of expected developments in the foreground and background systems ([Table 1](#)). This framework consists of three phases: (I) definition of the development stage, (II) assessment of developments toward technology and manufacturing readiness and (III) assessment of developments for industrially produced technologies. A comprehensive description of its application is provided in section 3. of Supplementary Materials 1. Process changes and process synergies were derived from reports of the Dutch ‘Manufacturing Industry Decarbonisation Data Exchange Network’ ([PBL, 2021](#)). An overview of all identified developments and justification for inclusion of select developments in our case study is provided in [Table S. 5.](#) of Supplementary Materials 1. Assessment of external developments in the electricity sector were included for which projections were derived from the integrated assessment model IMAGE ([Stehfest et al., 2014](#)). Assessment of Shared Socioeconomic Pathway 2

Table 1

Application of the prospective LCA framework in van der Hulst et al. (2020) to the case study of chemical recycling of feedstock using the Synova/T.EN system. TRL: Technology readiness level, expressed on a scale from 1 to 9 (see Mankins (1995)); MPL: market penetration level, expresses the percentage market share of a technology; SSP2: shared socioeconomic pathways 2 'Middle of the road' narrative; RCP4.5: representative concentration pathway 4.5 W/m² by 2100.

Phase: Step	Modelled change	Current state (2020)	Future state (2030)
Phase I: Definition of development stage	Technological development	TRL 6 Incorporation in production site (TRL7) is being explored (Buchner et al., 2019)	TRL 9; MPL 13% projected global MPL for chemical recycling routes like the one described herein when considering scenarios that aim for a 50% reuse and recycling rate of plastics (Hundertmark et al., 2018)
Phase II: Process changes	<ul style="list-style-type: none"> Decarbonization of steam production Side product valorisation 	<ul style="list-style-type: none"> Steam generation in a conventional boiler which uses fuel gas Fuel gas side-product is used as energy source in steam production and post-processing and a surplus is used for heat and power co-generation 	<ul style="list-style-type: none"> Two options for decarbonisation of steam production were assessed (PBL, 2021): <ul style="list-style-type: none"> Steam production in an electric boiler Steam production in a biogas boiler All fuel gas is converted to syngas in steam reforming and subsequently converted to methanol in methanol synthesis (Althaus et al., 2007). Methanol is converted to olefins in methanol-to-olefins synthesis (Rosental et al., 2020)
Phase II: Size scaling	Equipment scaling	Equipment with a throughput of 150 kg feedstock/h	Equipment with a throughput of 6376 kg feedstock/h. Inventory flows were observed to scale approximately linearly with feedstock throughput for size scaling of equipment from 5 kg/h (lab scale) to 150 kg/h (pilot scale) and even 1200 kg/h (demonstration scale) throughput, assuming feedstock of comparable composition and unchanged residence times. Since the functional unit is formulated in terms of feedstock throughput, interventions per kg of feedstock treated remain the same for linear size scaling of equipment.
Phase II: Process synergies	Waste valorisation	Captured carbon dioxide from product gas cleaning is emitted to the atmosphere	Captured carbon dioxide is converted to methanol in methanol synthesis using hydrogen obtained through alkaline water electrolysis using Dutch grid mix average electricity. Methanol is subsequently converted to olefins in methanol-to-olefins (MTO) synthesis (Rosental et al., 2020)
Phase III: Industrial learning	Not applicable	System is not yet applied at an industrial scale	Industrial application of the Synova/T.EN system in 2030 were assumed to be too limited for substantial industrial learning to occur.
Phase III: External developments	Decarbonization of electricity production	Electricity market of 2020 (ecoinvent, 2020)	Electricity market of 2030 (Mendoza Beltran et al., 2020). Two narratives for the development of the electricity market were assessed: <ul style="list-style-type: none"> SSP2 baseline SSP2 RCP4.5

(SSP2) was conducted, in which social, economic, and technological trends are expected to follow historic trends (O'Neill et al., 2014). The baseline scenario from this narrative was assessed, as well as the representative concentration pathway 4.5 W/m² (RCP4.5) (van Vuuren et al., 2011). The baseline scenario represents a world with no concerted international effort to address climate change, while the RCP4.5 scenario assumes efforts towards intermediate GHG emissions, resulting in an estimated global warming in 2100 between 2.1 and 3.5 °C. Electricity market datasets in ecoinvent 3.7 were adapted using the approach in Mendoza Beltran et al. (2020) to reflect projected production mix compositions for 2030.

GHG benefits of developments in Table 1 were assessed in five distinct scenarios (Table 2). The current scenario (Fig. 2b) describes chemical recycling and incineration in year 2020, while Future scenarios 1–4 describe year 2030. Future 1–3 describe specific sets of

developments in Table 1, which are decarbonization of steam production (Future 1; blue in Fig. 2c), side product and waste valorisation (Future 2; green in Fig. 2c) and decarbonisation of electricity production (Future 3; purple in Fig. 2c). Futures 1 and 3 contain two sub-scenarios to assess different development options. Future 4 describes all developments combined and contains four sub-scenarios.

2.4. GHG footprint calculations

Following the IPCC 2013 method (Intergovernmental Panel on Climate Change, 2014), Global Warming Potentials (GWPs) for a 100-year time horizon and excluding carbon-climate feedbacks were used to quantify the GHG footprint. Here, this method is adapted to allow for negative emissions from technologies introduced to the grid mix in the future such as bioenergy with carbon capture and storage

Table 2

Overview of the assumptions for each scenario assessed in this work. CHP: combined heat and power production; SSP2: shared socioeconomic pathways 2 'Middle of the road' narrative; RCP4.5: representative concentration pathway 4.5 W/m² by 2100; EB: electric boiler; BB: biogas boiler; BL: baseline narrative of SSP2; RCP: RCP4.5 narrative of SSP2.

Scenario	Current	Future 1		Future 2	Future 3		Future 4			
		EB	BB		BL	RCP	EB-BL	BB-BL	EB-RCP	BB-RCP
Sub-scenario										
Steam production										
Fuel gas boiler	X			X	X	X				
Electric boiler		X					X		X	
Biogas boiler			X					X		X
Fate of side-product and waste										
Carbon dioxide venting to air & fuel gas CHP	X	X	X		X	X				
Olefins from fuel gas & captured carbon dioxide				X			X	X	X	X
Electricity production										
2020 market	X	X	X	X						
2030 market, SSP2 baseline narrative					X		X	X		
2030 market, SSP2 RCP4.5 narrative						X			X	X

(BECCS). A description of method adaptations and a complete list of resulting GWPs is provided in section 3. of Supplementary Materials 1.

2.5. Sensitivity analyses

Assessment of systems with multiple functions can be conducted using various approach. While standards like ISO 14040 prioritized these solutions for multifunctionality in a fixed hierarchy, the selected solution should in the first place correspond to goal and scope definitions (Schaubroeck et al., 2022). Considering the comparative goal of our study, system expansion was considered a solution that is equally as valid as the method of substitution selected in the goal and scope. section 4. of Supplementary Materials 1, results were reproduced using the system expansion method to assess the sensitivity of results to this modelling choice. In both solutions to multifunctionality, conventional products must be selected, either to represent what products is replaced in the case of substitution or what additional products is required in the case of system expansion. The choice of this conventional product is uncertain (Hanssen & Huijbregts, 2019). In section 5. of Supplementary Materials 1, a sensitivity analysis was conducted to assess what results would be obtained when selecting marginal production for the conventional products, rather than market average production.

3. Results

3.1. Current situation

Diverting post-consumer mixed plastic waste from incineration to chemical recycling resulted in a GHG benefit of 0.82 kg CO₂-eq./kg feedstock treated in year 2020 (Fig. 3). For incineration, GHG savings from energy recovery are not enough to offset direct GHG emissions from the incineration process, resulting in a net positive GHG footprint of 0.78 kg CO₂-eq./kg feedstock treated. For chemical recycling, GHG emissions are more or less offset by GHG savings from obtained energy and HVC products, giving chemical recycling a net-negative GHG footprint of -0.04 kg CO₂-eq./kg feedstock treated. Direct CO₂ emissions in the form of flue gas from char and tar combustion, venting of CO₂ removed in the gas cleaning stage, and indirect GHG emissions from electricity consumption were the main contributors to the GHG footprint of the Synova/T.EN system.

3.2. Future scenarios

The current GHG benefit of 0.82 kg CO₂-eq./kg feedstock treated can increase by 67% to as much as 1.37 kg CO₂-eq./kg feedstock or decrease by 65% to as little as 0.29 kg CO₂-eq./kg feedstock, depending on the assessed future scenario (Fig. 4). The incumbent technology of incineration with energy recovery is only affected by developments in electricity generation. Therefore, results for chemical recycling in scenarios Future 1 and 2 are compared to the current scenario for incineration, whereas results for scenarios Future 3 and 4 are compared to corresponding future scenarios for incineration. This is indicated with different dashed lines.

Combining all developments (Future 4) results in the largest GHG benefits, in particular for the scenarios in which decarbonization of the electricity sector follows a more progressive development pathway (1.33 and 1.37 kg CO₂-eq./kg feedstock treated for Future 4-EB-RCP and Future 4-BB-RCP respectively).

3.3. Sensitivity analyses

Presented results were reproduced using system expansion instead of substitution as solution to multifunctionality (see section 4. of Supplementary Materials 1). Net GHG footprints for incineration remain more or less constant over time at around 4.25 kg CO₂-eq./kg waste feedstock treated. Net GHG footprints for chemical recycling differ per scenario, ranging from 2.84 to 3.95 kg CO₂-eq./kg waste feedstock treated. While these net GHG footprints are considerably larger than those found using the substitution method due to differences in accounting for products, the relative difference between both product systems remained the same, i.e., the GHG benefits were found to be equivalent. However, changes to GHG benefits were observed when selecting marginal production for avoided products instead of market average production (see section 5. of Supplementary Materials 1). For 2020, selecting the marginal suppliers instead of the grid mix average decreased GHG benefits by 5% to 0.78 kg CO₂-eq./kg feedstock treated, while for 2030 GHG benefits increased by 16–31% to 1.41–1.59 kg CO₂-eq./kg feedstock treated when considering all developments combined (i.e., Future 4).

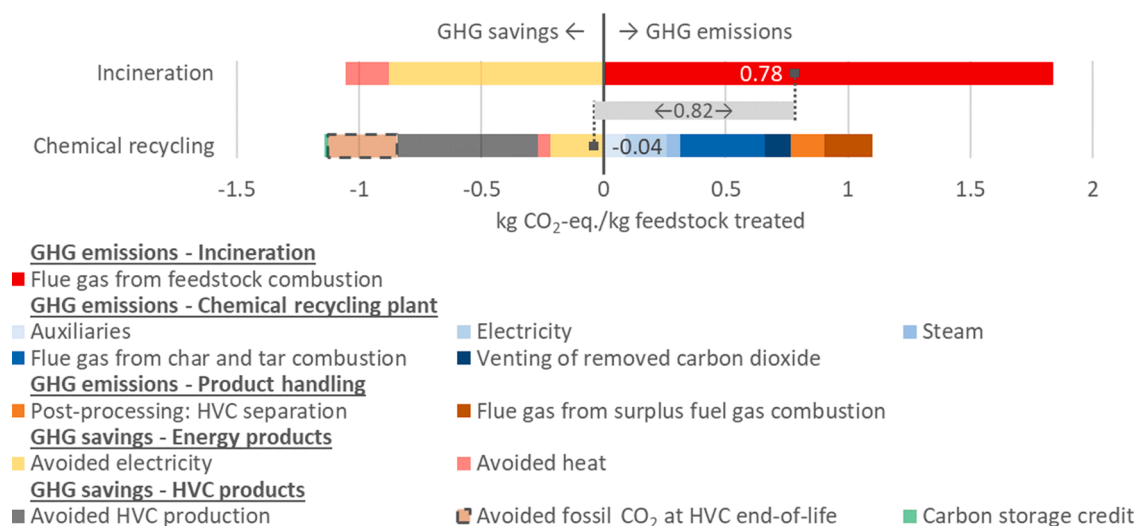


Fig. 3. Process contribution analyses of GHG footprints for treatment of 1 kg feedstock in year 2020. Numerical values are provided in Supplementary Materials 2. Net GHG footprints for each scenario are indicated with a grey square. Emission saving potential is indicated with a grey bar. The GHG savings from avoided fossil CO₂ emissions at HVC end-of-life are marked with a dashed border. These savings are to be subtracted from the results in case one assumes a use and end-of-life scenario for the HVCs or products derived in which incineration is not the end-of-life fate. GHG: greenhouse gas; CO₂: carbon dioxide; HVC: high value chemical.

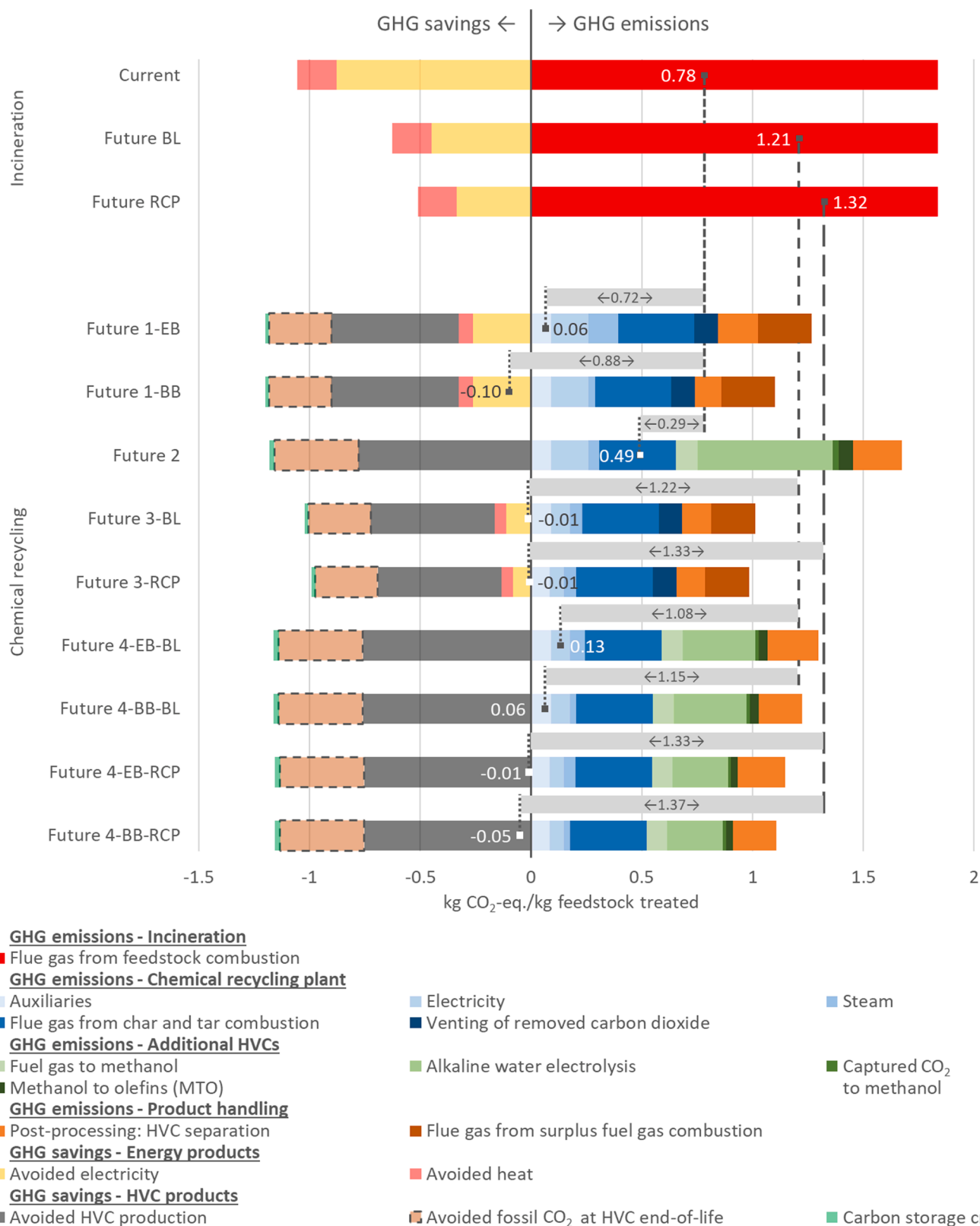


Fig. 4. Process contribution analyses of GHG footprints for treatment of 1 kg in year 2030. Numerical values are provided in Supporting Materials 2. Net GHG footprints for each scenario are indicated with grey/white squares. Emission saving potentials are indicated with grey bars. The GHG savings from avoided fossil CO₂ emissions at HVC end-of-life are marked with a dashed border. These savings are to be subtracted from the results in case one assumes a use and end-of-life scenario for the HVCs or products derived in which incineration is not the end-of-life fate. GHG: greenhouse gas; BL: baseline scenario; RCP: representative concentration pathway 4.5 W/m² by 2100 scenario; EB: electric boiler; BB: biogas boiler; CO₂: carbon dioxide; HVC: high value chemical.

4. Discussion

4.1. Interpretation

Direct emissions in both the chemical recycling and incineration processes contributed most to the GHG footprints. In incineration, all carbon in the feedstock is oxidized to carbon dioxide and only energy products are obtained, giving incineration a carbon efficiency of 0%. Chemical recycling has fewer direct emissions and more indirect emissions compared to incineration. Part of the feedstock is retained in a reduced state as aliphatic chemical compounds, giving the Synova/T.EN system a carbon efficiency of 53% (see section 3. of Supplementary Materials 1). While indirect emissions from consumed energy and materials are larger for chemical recycling compared to incineration, these emissions were more than offset by reduced direct emissions and larger avoided emissions from producing HVCs instead of energy products. Thus, retention of carbon in the technosphere through chemical recycling results in GHG benefits when compared to the incumbent technology where all carbon is lost to the atmosphere.

In scenario analyses for 2030, we found that decarbonization of steam production has little influence on the GHG footprint of chemical recycling. Relative to the current situation (-0.04 kg CO₂-eq./kg feedstock treated, [Figure 3](#)), the net GHG footprint decreased by 0.06 kg CO₂-eq./kg feedstock treated when utilizing a biogas boiler (Future 1-BB), while it increased by 0.10 kg CO₂-eq./kg feedstock treated when using an electric boiler (Future 1-EB). Assumed boiler efficiencies ranged from 86% for a fuel gas boiler, 88.5% for a biogas boiler and 99.9% for an electric boiler ([PBL, 2021](#)). While the electric boiler is most energy efficient, the additional conversion of fuel to electricity reduces the overall efficiency, which explains the larger GHG footprint.

Utilization of fuel gas and captured carbon dioxide (Future 2) was calculated to increase the carbon efficiency of the Synova/T.EN system to 71% (see section 3. of Supplementary Materials 1). While this further reduces indirect emissions of the chemical recycling process, it more strongly increases indirect emissions. We only considered utilization of carbon dioxide removed from the product gas mixture, since this is an integral and essential part of the Synova/T.EN system and, therefore, removed carbon dioxide is available at no additional cost, contrary to carbon dioxide present in e.g., flue gasses. While there are many pathways for carbon capture and utilization (CCU) available ([de Kleijne et al., 2022](#)), we limited ourselves to utilization in production of additional olefins *via* methanol synthesis and MTO. Methanol synthesis requires an input of externally sourced hydrogen, which is conventionally produced through steam reforming of natural gas, referred to as grey hydrogen. However, we assumed hydrogen production through alkaline water electrolysis, since this is more in line with other assessed decarbonization goals. The large consumption of electricity in alkaline water electrolysis is a major contributor to the GHG footprint of this scenario, resulting in an increased net GHG footprints and therefore reduced GHG benefits relative to incineration. Utilization of fuel gas and captured carbon dioxide would therefore only be recommended when indirect emissions are simultaneously reduced. With the current emission factor for Dutch electricity, the GHG footprint of alkaline water electrolysis would be 31.4 kg CO₂-eq./kg H₂. This reduces to 16.8 and 12.9 kg CO₂-eq./kg H₂ in 2030 for the baseline and RCP 4.5 development pathways respectively. As a result, the best case GHG footprint for alkaline water electrolysis in 2030 is still larger than that of steam reforming of natural gas, which is approximately 11.5 kg CO₂-eq./kg H₂ ([Bauer et al., 2022](#)). However, when using dedicated renewable electricity, green hydrogen could be produced, with a very low carbon footprint of ≤ 1 kg CO₂-eq./kg H₂ ([Hydrogen Council, 2021](#)), which would make conversion of captured carbon dioxide a no-regret option.

The future change in the carbon intensity of electricity production has important consequences for the GHG footprint of incinerating the post-consumer mixed plastic waste. Incineration with energy recovery is a net electricity producer, for which it receives credits through avoiding

the production of grid mix average electricity. The emission factor of Dutch electricity was found to reduce from 0.60 kg CO₂-eq./kWh in 2020 to 0.31 or 0.23 kg CO₂-eq./kWh in 2030, assuming business-as-usual (Future 3-BL) or more progressive (Future 3-RCP) development pathways for the electricity sector respectively. This reduction in emission factor negatively impacts the GHG footprint for incineration: credits from avoided electricity diminish and net GHG footprints increase. On the other hand, chemical recycling consumes about as much electricity as it produces and therefore electricity grid mix decarbonization has little effect on the GHG footprint. This effect becomes more pronounced when considering other future developments. Electrification of steam production (Future 1-EB) and CCU with hydrogen from electrolysis (Future 2) both increase electricity consumption, while conversion of fuel gas to HVCs (Future 2) decreases electricity production by the chemical recycling system, making chemical recycling a net electricity consumer. When considered in isolation, these developments lead to increased GHG footprints for chemical recycling. However, combining these developments with the decarbonization of the electricity grid mix results in equivalent GHG footprints (Future 4-EB-BL and Future 4-EB-EB-RCP) and GHG benefits that are larger than that of the current system. This highlights the importance of electricity grid mix decarbonization in creating GHG benefits from chemical recycling of plastic waste.

Assessing sensitivity of presented results to modelling choices revealed that while different valid solutions to multifunctionality resulted in different net GHG footprints, the net GHG benefits remained the same. Substitution and system expansion are considered mathematically equivalent for comparative LCAs. However, the choice in avoided products did have an influence on GHG benefits, with marginal suppliers providing larger GHG benefits over the long run. Nonetheless, general conclusions were unaffected: diverting plastic waste from incineration to chemical recycling provides GHG benefits which will increase due to future developments.

Emission saving potentials of the chemical recycling system were reported relative to incineration with energy recovery. Changes to the foreground system of this incumbent, such as efficiency improvements or emission reductions, were not considered, which could result in an overestimation of the GHG footprint of incineration. Furthermore, application of carbon capture and storage was not considered, because its implementation is currently too uncertain. While incineration with energy recovery is the dominant treatment method for the Netherlands and many other European countries, landfilling is the dominant option in other countries ([PlasticsEurope, 2020](#)). Landfilling can act as carbon sink for the plastic fraction in the feedstock but can also result in emissions of methane from decomposition of the biogenic fraction in the feedstock under anaerobic conditions. The net GHG footprint for landfilling of the studied feedstock is unknown. In addition to incineration and landfilling, plastic waste is increasingly treated through mechanical recycling ([PlasticsEurope, 2020](#)), which has a GHG footprint that is comparable to or lower than that of chemical recycling ([Davidson et al., 2021](#)). However, mechanical recycling is unfit for treatment of waste that is very heterogeneous and contaminated with non-plastic components, as is the case for the feedstock considered in our study.

To the authors knowledge, the Synova/T.EN system is the only system assessed in LCA literature that performs direct chemical recycling of post-consumer mixed plastic waste to olefins. A comparison of our case study to the screening LCA of this system reported by [Uijtewaal and Broeren \(2021\)](#) is included in section 6. of Supplementary Materials 1. Other systems assessed in LCA literature convert plastic waste to either syngas or a naphtha-like pyrolysis oil. Comparison to such alternative chemical recycling systems should be made with caution, since GHG footprints are influenced by waste feedstock composition and amount and type of products obtained. Emission saving potentials presented herein are therefore only representative for the Synova/T.EN chemical recycling system with a distinct feedstock as waste input.

4.2. Recommendations for future research

Efforts to further reduce GHG footprints for chemical recycling with the Synova/T.EN system should be focussed on major contributors that were identified with contribution analyses. Direct emissions from combustion of chars and tars could be reduced by scrubbing carbon dioxide from the flue gasses and subsequently processing it together with captured carbon dioxide from product gas cleaning. Direct emissions from fuel gas consumption in post-processing could be reduced by employing furnaces that use biogas or electricity as energy source. Indirect emissions from electricity consumption in green hydrogen production could be reduced by using dedicated renewable energy sources or by utilizing alternative production routes based on e.g., biomass. Whether these additional emissions reduction options would be technologically and economically feasible remains to be assessed.

For future trials with the Synova/T.EN system, it is recommended to either evaluate the polymer composition of the feedstock prior to conversion or use a feedstock of known polymer composition. Resulting data could then also be used to potentially infer relationships between the composition of feedstock and obtained HVCs. This could enable dynamic GHG footprint assessment of feedstock with varying composition, allowing for comparison to studies with alternative feedstock compositions.

A more comprehensive assessment of emissions from the product system is recommended to improve the impact assessment. Assessment of fugitive methane emissions from the production, distribution and conversion of fuel gas would be relevant, since these could significantly increase GHG footprints for the chemical recycling system, thereby reducing emission savings potentials (Layritz et al., 2021). Furthermore, evaluation of non-GHG emissions could enable assessment of other impact categories to identify potential trade-offs in diverting plastic waste from incineration to chemical recycling. The studies of BASF (2020) and Jeswani et al. (2021), for example, found trade-offs with the impact categories acidification, freshwater and marine eutrophication, photochemical ozone formation and human toxicity, meaning the impact in these categories was larger for their chemical recycling system than for incineration.

5. Conclusion

Direct chemical recycling of post-consumer mixed plastic wastes to olefins with the Synova/T.EN system in 2020 was found to result in a GHG benefit of 0.82 kg CO₂-eq./kg waste feedstock treated when compared to waste incineration with energy recovery. This GHG benefit was found to increase to as much as 1.37 kg CO₂-eq./kg waste feedstock treated by 2030 when combining the effect of decarbonization of steam and electricity production and process optimizations to increase carbon efficiency from 53 to 71% through conversion of side product fuel gas and captured carbon dioxide to olefins. Meanwhile, electricity decarbonization increased the GHG footprint of plastic waste incineration with energy recovery due to diminishing credits for avoided electricity production. Our findings therefore imply that GHG benefits from direct chemical recycling of post-consumer mixed plastic waste will only increase in a decarbonizing economy.

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CRedit authorship contribution statement

Mitchell K. van der Hulst: Conceptualization, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft. **Anne B. Ottenbros:** Formal analysis, Investigation, Writing –

review & editing. **Bram van der Drift:** Conceptualization, Resources, Writing – review & editing. **Špela Ferjan:** Formal analysis, Investigation. **Toon van Harmelen:** Conceptualization, Supervision. **Anna E. Schwarz:** Conceptualization, Writing – review & editing. **Ernst Worrell:** Supervision. **Rosalie van Zelm:** Conceptualization, Supervision, Writing – review & editing. **Mark A.J. Huijbregts:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing. **Mara Hauck:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. Editorial Board Member of Resources, Conservation & Recycling: Ernst Worrell.

Data Availability

The data that has been used is provided in the Supplementary Materials, excluding any confidential data.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2022.106582. This work contains two electronic files with supplementary materials. Supplementary Materials 1 is a PDF containing a table on technology providers of chemical recycling, a description of feedstock composition, a comprehensive description of the materials and methods, sensitivity analyses, and a comparison of results to Uijtewaal and Broeren (2021). Supplementary Materials 2 is a spreadsheet containing the numerical values for results presented in Fig. 3 and Fig. 4, as well as the raw data and calculations for unit processes and emissions factors used to obtain GHG footprints of the product system for the various assessed scenarios.

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